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A Superconducting Detector System for High-Resolution Energy-Dispersive Soft X-Ray Spectroscopy

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DISCLAIMER

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INTRODUCTION

Synchrotron-based soft x-ray spectroscopy is often limited by detector performance. Grating spectrometers have the resolution, but lack the efficiency for the analysis of dilute samples. Semiconducting Si(Li) or Ge detectors are efficient, but often lack the resolution to separate weak signals from strong nearby lines in multi-element samples. Superconducting tunnel junctions (STJs) operated at temperatures below 1 K can be used as high-resolution high-efficiency x-ray detectors. They combine high energy resolution around 10 eV FWHM with the broad band efficiency of energy-dispersive detectors. We have designed a two-stage adiabatic demagnetization refrigerator (ADR) to operate STJ detectors in x-ray fluorescence measurements at beam line 4 of the ALS. We demonstrate the capabilities of such a detector system for fluorescence analysis of dilute metal sites in proteins and inorganic model compounds.

THE DETECTOR SYSTEM

Superconducting tunnel junction (STJ) x-ray detectors consist of two superconducting electrodes separated by a thin insulating barrier. They are based on measuring the excess tunneling current after x-ray absorption in one of the electrodes generates excess free charge carriers in proportion to the x-ray energy. In typical Nb-based STJ detectors, statistical fluctuations in the charge generation and tunneling processes limit the energy resolution to 4.3 eV FWHM for 1 keV photons. Their maximum pile-up limited count rate is determined by the life time of the excess charges of several µs. Over the last decade, the Advanced Detector Group at Lawrence Livermore National Laboratory (LLNL) has been developing Nb-Al-AlOx-Al-Nb STJ detectors for x-ray astrophysics and national security applications. They have achieved an energy resolution between 1.7 eV and 8.9 eV FWHM for photon energies between 50 eV and 1 keV, and they can be operated at count rates in excess of 10,000 counts/s [1, 2].

In collaboration with the Advanced Detector Group at LLNL, we have developed a two-stage adiabatic demagnetization refrigerator (ADR) to operate STJ detectors in synchrotron-based x-ray fluorescence measurements (figure 1) [3]. Adiabatic demagnetization is a process of magnetic cooling below the temperature of a liquid helium bath by isothermal magnetization and adiabatic demagnetization of a paramagnetic material. Our ADR uses two different paramagnets. The first stage is cooled by a gallium gadolinium garnet (GGG) to 1 K and supports a second stage cooled by a ferric ammonium alum (FAA) paramagnet. This two-stage design attains a 60 mK base temperature with 16 hours hold time per demagnetization cycle. It is reliable, easy to operate and does not require pumping on the liquid helium bath.

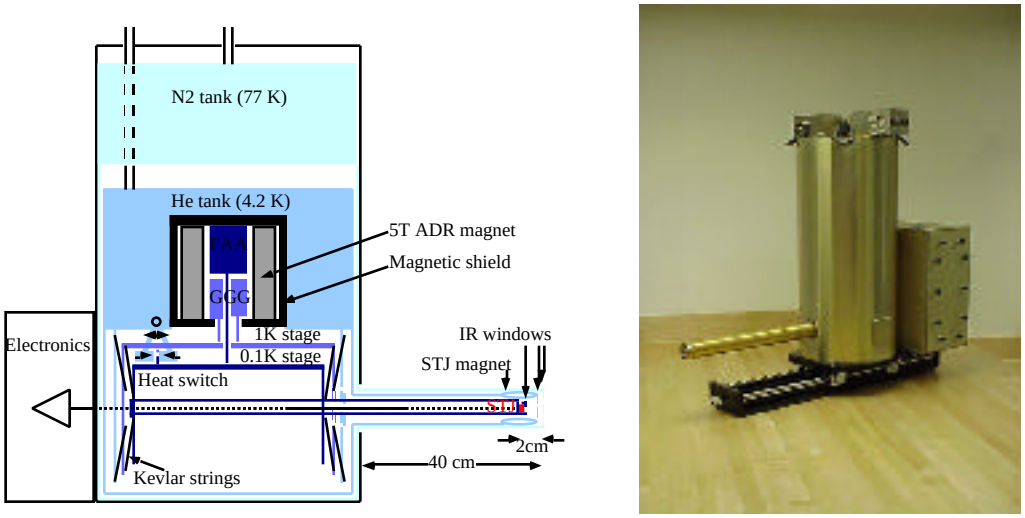

Figure 1. Schematic and photograph of the ADR cryostat. The STJ detector operates at a temperature of 0.1 K at the end of the detector snout that can be inserted into a UHV chamber.

To adapt this cryostat for x-ray fluorescence measurements, the STJ detector is held at the end of a 40 cm long cold finger that can be inserted into a sample chamber. A liquid He cooled and a liquid N_2 cooled shield allow operation of the STJ detector behind three thin IR blocking windows within 2 cm of a room temperature sample in a UHV chamber at a pressure in the low 10^{-9} Torr range. The x-ray induced signal is read out with a custom-designed FET preamplifier at room temperature followed by standard commercial pulse processing electronics.

X-RAY FLUORESCENCE MEASUREMENTS

We are interested in fluorescence detected absorption spectroscopy of transition metal L-edges in metalloenzymes and corresponding model compounds. Absorption spectra are taken by scanning the energy of the incident monochromatized synchrotron beam through an L-edge of interest to directly probe the empty states in the 3d manifold. Since these 3d levels are involved in chemical binding, absorption spectroscopy provides information about the metal oxidation state and the ligands, which in turn can be related to the catalytic mechanisms of the enzyme. For dilute metalloenzyme samples, the sensitivity of the absorption spectroscopy is enhanced by measuring the intensity of the corresponding L fluorescence line. STJ detectors are used to efficiently separate this fluorescence signal from nearby emission lines in cases where Ge detectors lack the energy resolution and grating spectrometers lack the detection efficiency.

Initial x-ray fluorescence tests of the STJ detector system were performed at beam line 4.0.2. at the ALS. Figure 2a shows a fluorescence spectrum of the metalloprotein hydrogenase of the bacterium *D. Gigas* containing ≈480 ppm nickel and ≈5800 ppm iron. The 2-hour spectrum was acquired with a 100 µm × 100 µm STJ with a detector-sample distance of ≈25 mm. While the detector resolution between 11 and 25 eV FWHM for the energy range between 277 eV (C K) and 850 eV (Ni L) is still somewhat less than that of nominally identical detectors operated in an older cryostat without a snout [1], it is more than sufficient to separate the Fe L and the Ni L signal from the large C K and O K background. For comparison, we have included a fluorescence spectrum of the same protein taken with a commercial 30-element Ge detector.

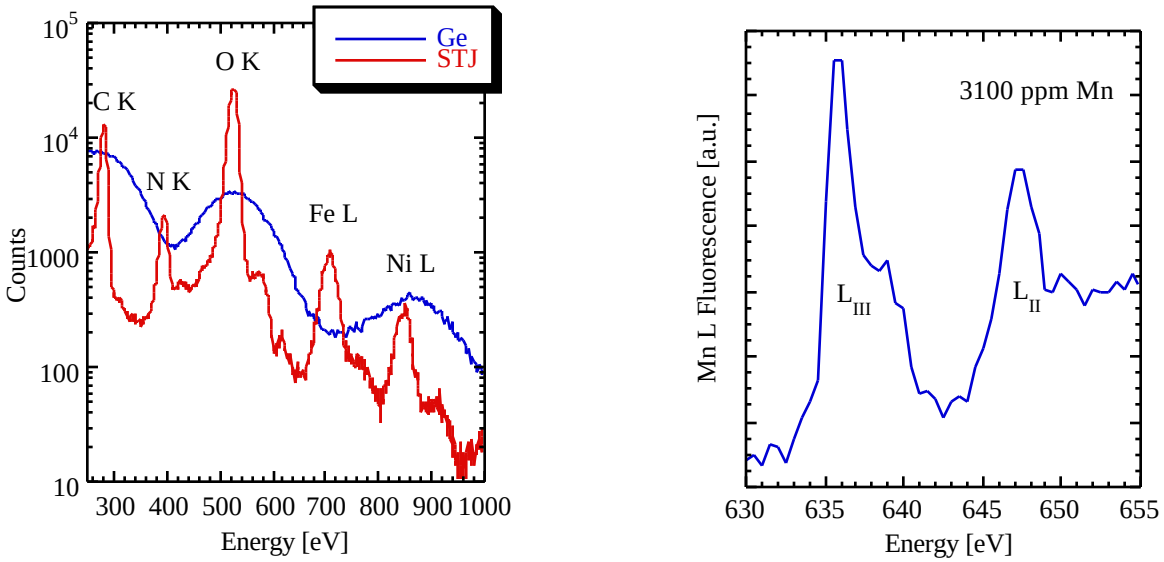

Figure 2a (left): Soft X-ray emission spectra of the metalloprotein hydrogenase containing about 480 ppm Ni and 5800 ppm Fe taken with the STJ detector system (red) and, for comparison, with a commercial 30-element Ge detector (blue). The Ni line is enhance in the Ge detector spectrum because of resonant excitation at the Ni L edge. Figure 2b (right): Fluorescence-detected L-edge absorption spectrum of 3100 ppm Mn in an MgO crystal.

The high energy resolution of STJ detectors allows fluorescence-detected absorption spectroscopy of dilute metals even in the presence of strong nearby emission lines. The fluorescence-detected absorption spectrum of Mn impurities in an MgO crystal in figure 2b provides an illustrative example. It was taken with a single 200 µm × 200 µm STJ in ≈1 hour and is not affected by the O K fluorescence at 525 eV. The spectra demonstrate how cryogenic STJ detectors can greatly enhance the sensitivity in soft X-ray spectroscopy of dilute samples.

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